STUDY OF THERMAL PROPERTIES OF REFRACTORIES
(FIRST QUARTERLY PROGRESS REPORT)

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Contract No. AF 33(657)-7136

ARPA Order No. 24-61

Project No. 002

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October 1961

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Canoga Park, California

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Abstract

A status report is presented on the development of the transient thermal property apparatus described in Parts I and II of WADD-TR-60-581. Modifications and improvements of the apparatus are described, and results of measurements of the thermal diffusivity of tantalum at 1400° and 1500°C are given. The status of thermal property measurements by existing techniques is also included.

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Foreword

The work reported here was performed by Atomics International, a Division of North American Aviation, under the auspices of the Department of Defense through the Advanced Research Project Agency. Contract AF 33(657)-7136 issued under ARPA Order No. 24-61, Project 002, "Materials Thermal Properties", was administered by the Directorate of Materials and Processes, Deputy for Technology, Aeronautical Systems Division, with Mr. Hyman Marcus acting as project engineer. This report covers work conducted from August 1, 1961 to September 30, 1961.

This investigation is an extension of work previously performed under Contract AF 33(616)-6794 which was under the responsible supervision of Dr. N. R. Mukherjee of the Thermoelectric and High Temperature Materials Section. The present work is under Dr. S. C. Carniglia of the Fuels and Materials Department. Mr. Mark Nakata has joined in this work and has assumed an important role in the experimental phase of the transient thermal diffusivity apparatus.

Study of Thermal Properties of Refractories

by R. E. Taylor and M. M. Nakata

I. Introduction

The primary purpose of this project is to measure, describe, and understand the thermal properties of refractory materials from ambient temperature to temperatures approaching the melting point. This quarterly progress report covers the period from August 1, 1961 to September 30, 1961.

Due to delays in funding, work was carried out at a low level of activity during most of the quarter. However, because essentially all of the developmental work on the photoelectric pyrometer had been completed during the preceding period, substantial progress was made toward the development of the apparatus for transient thermal property measurements. This phase of the work is described in Section II.

In order to obtain information on the thermal properties of refractory materials during the development of new approaches, an effort utilizing existing techniques has been maintained. The status of this work is described in Sections III and IV.

II. Status Report on Transient Thermal Property Apparatus

The increased sensitivity resulting from the completion of the photoelectric pyrometer described in past reports[1,2] and the use of photographic techniques pinpointed problems in shielding and sample holes. Specifically, it was discovered that a significant amount of stray radiation was entering the detector and that the focusing arrangement was inadequate.

Modifications in the radiation shield assembly were therefore made, 1) to reduce the amount of heat generated in the shields due to RF energy pickup, and 2) to cut down the amount of spurious radiant energy reaching the detector to a negligible level, by means of suitable shields and collimators. The configuration is shown in Figure 1. A marked decrease in RF energy pickup by the tantalum shields was obtained by placing them just above the concentrator instead of extending them into the space occupied by the heater and specimen. Further reduction in the shield temperature was made by cutting radial slits and punching pinholes in the shields.

The major source of spurious radiation was found to be the scattered light emanating from the space between the individual shields and between the shield assembly and the sample. Ceramic thermocouple tubes approximately .060 in. I.D. and 1/2 in. long were inserted through the sight holes of the shields to within

approximately 0.01 in. from the sample, to correct this condition. It was also found that the diffuse radiation from the outermost shield and scattered light from the pyrex vacuum enclosure contributed a significant amount of radiation to the detector. A secondary radiation shield consisting of an alumina ring which encircles the tantalum shields, and an alumina disc which fits on the ring, all but eliminated the diffuse radiation reaching the pyrometer. Finally, a small but significant amount of radiation was being picked up by the pyrometer from the sight hole which was not being measured, because of the proximity of the holes and the distance of the pyrometer from the specimen. A sheet of metal in which two 0.1 in. holes were drilled to match the sight holes, was placed over the sight windows, so that the narrow beam of radiation from only the sight hole being measured would reach the pyrometer optics.

Two 0.060 in. sight holes were drilled into the specimen to a depth of 7/16 in., and concentric holes, 0.040 in., were drilled within the larger holes to a depth of 1/8 in. The narrow shoulder at the bottom of the larger holes was then used as a guide to focus the pyrometers accurately on the bottom of the sight holes. The radiant energy passing to the phototube was limited to an area of approximately 0.030 in. diameter by adjusting an iris diaphragm in the optical system.

The photoelectric pyrometer was described in a previous

report.[2] Minor adjustments were made to improve the focusing mechanism and to prevent changes in the photomultiplier output due to slight shifting of the phototube within the pyrometer housing.

Improvements were made in the specimen and heater supports so that inconsistencies in the pattern of thermal transport which were observed in some of the previous runs were completely eliminated.

Absolute temperature determinations were made with an optical pyrometer. The pyrometer used was a Pyro Mico optical pyrometer, which was calibrated against a tungsten filament lamp calibrated at the National Bureau of Standards. The maximum uncertainty in the present calibration is estimated to be ± 17°C. The repeatability of readings for this pyrometer is ± 2°C. Correction for the absorption of the sight window and prism which were used in conjunction with the optical pyrometer was also determined.

The photocurrent output from each of the sight holes at steady state was measured and plotted against the temperatures observed with the optical pyrometer to determine whether the photocurrent output was an accurate function of the temperature of the holes. The calibration curve obtained in the present work is shown in Figure 2. It is encouraging to note that,

within the sensitivity of the optical pyrometer, the photocurrent output is the same for both sight holes, over the temperature range covered. This is an indication that the contribution of spurious radiation has been reduced to an insignificant level. Although the optical pyrometer will indicate a temperature change of ± 2°C (whereas the sensitivity of the photoelectric pyrometer is an order of magnitude greater), if sufficient calibration points are recorded, a reasonably accurate calibration can be obtained by means of curve fitting.

a. Experimental Results

Tantalum was chosen as the first sample for transient thermal diffusivity determination. The high temperature thermal conductivity and specific heat of tantalum have been determined at this laboratory[3,4] so that the thermal diffusivity obtained experimentally can be compared with the values calculated from the equation:

$$\alpha = \frac{K}{cd} \quad ,$$

using these data.

The tantalum specimen was a right cylinder, 0.625 in. diam. by 1.375 in. long. The center sight hole was drilled at radius $r_1 = 0$ and the outer hole at $r_2 = \frac{a}{\sqrt{2}}$, where a is the radius of the sample. The thermal diffusivity calculated for tantalum is 0.212 cm^2/sec at 1500°C. When these values are used

in the following expression for our present specimen configuration,

$$\alpha = \frac{r_2^2 - r_1^2}{4\Delta t} = \frac{0.079}{\Delta t} ,$$

a Δt of only 0.37 sec. is indicated. Thus, tantalum is a difficult material to measure. On the other hand, if the system can be made to measure the thermal diffusivity of tantalum with sufficient accuracy, measurements of refractory compounds would then be relatively easy because their diffusivity is usually less than half that of tantalum.

Table I gives a comparison of the thermal diffusivity of tantalum measured with the present apparatus with the values calculated from the literature.

Table I Thermal Diffusivity of Tantalum

T °C	Liter. α cm^2/sec	Δt, sec Run 1	Run 2	Run 3	Aver.	Exp. α cm^2/sec
1400	0.211	0.35	0.33	0.35	0.34_3	0.230
1500	0.212	0.35	0.37	0.37	0.36_3	0.217

Considering the small Δt for tantalum, the reproducibility of the data is excellent. Diffusivity data between 1300° and 1400°C and between 1500° and 1700°C can also be determined with the present setup, but the temperature-time curves were not recorded in a

suitable form for accurate curve fitting in these temperature ranges. Data for these ranges will be given in a later report, but it may be stated here that values estimated by extrapolation agree within 20% of the literature values. Several avenues are still open for increasing the accuracy of the data. Multiplier phototubes with higher signal-to-noise response are now on the market and can be readily incorporated into our present pyrometer. Also, an alternate technique of covering a narrow temperature range per run is being investigated. This technique allows more accurate measurements in Δt by spreading out a narrow temperature range over the recorder chart.

III. Status Report on Steady State Thermal Conductivity Apparatus

Thermal conductivity measurements of specimens of zirconium carbide have been initiated and results will be given in the next report period.

The results of the thermal conductivity measurements of titanium carbide obtained in the laboratory[2] are in marked disagreement with that previously reported by Vasilos and Kingery[5]. Therefore, specimens of titanium carbide were cut from one of the samples measured in this laboratory and were sent to Dr. Michael Hoch of the University of Cincinnati for thermal conductivity measurements by another technique. It is expected that the results of Dr. Hoch will be available by the next reporting period.

IV. Status Report on Pulse Heating Technique for Specific Heat Determinations

The design of the sample holder for the specific heat determination by the pulse heating technique was improved over that previously described.[2] The major problems overcome by the new design are the ease of sample alignment and the development of voltage probes for use with samples where spot welding techniques are not satisfactory. The improved holder is shown in Figure 3. The sample shown mounted in the holder is titanium carbide. This sample was pulse heated to 1500°C several times with no evidence of failure. A heating rate of 6000°C per second was used and oscilloscopic traces of the voltage and current as a function of time were photographed. Therefore, specific heat measurements can be made on this type of brittle material. The major remaining problem is the lack of an apparatus for accurately determining the electrical resistivity of large (i.e., non-wire) samples at high temperatures. Developmental work on this apparatus is under way. In the meantime, data to 1000°C are being obtained and will be given in the next report.

The electrical resistivity of tungsten wire was measured from ambient to 1700°C. Tungsten/tungsten-26% rhenium thermocouple wire has been ordered to allow extension of the measurement range to at least 2300°C. After the measurement of the electrical resistivity has been extended to the higher temperature, the specific heat of tungsten will be obtained over the same temperature range.

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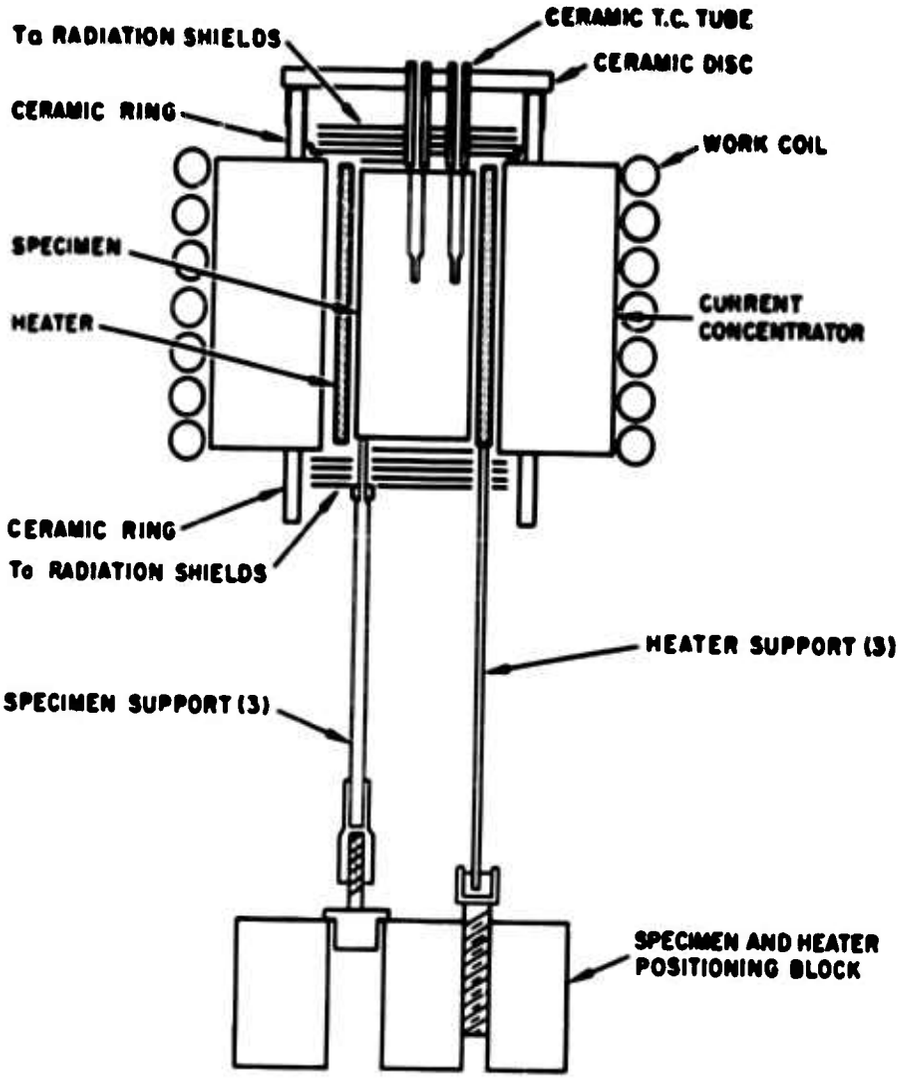

Figure 1. Cut-Out View of Transient Thermal Diffusivity Configuration

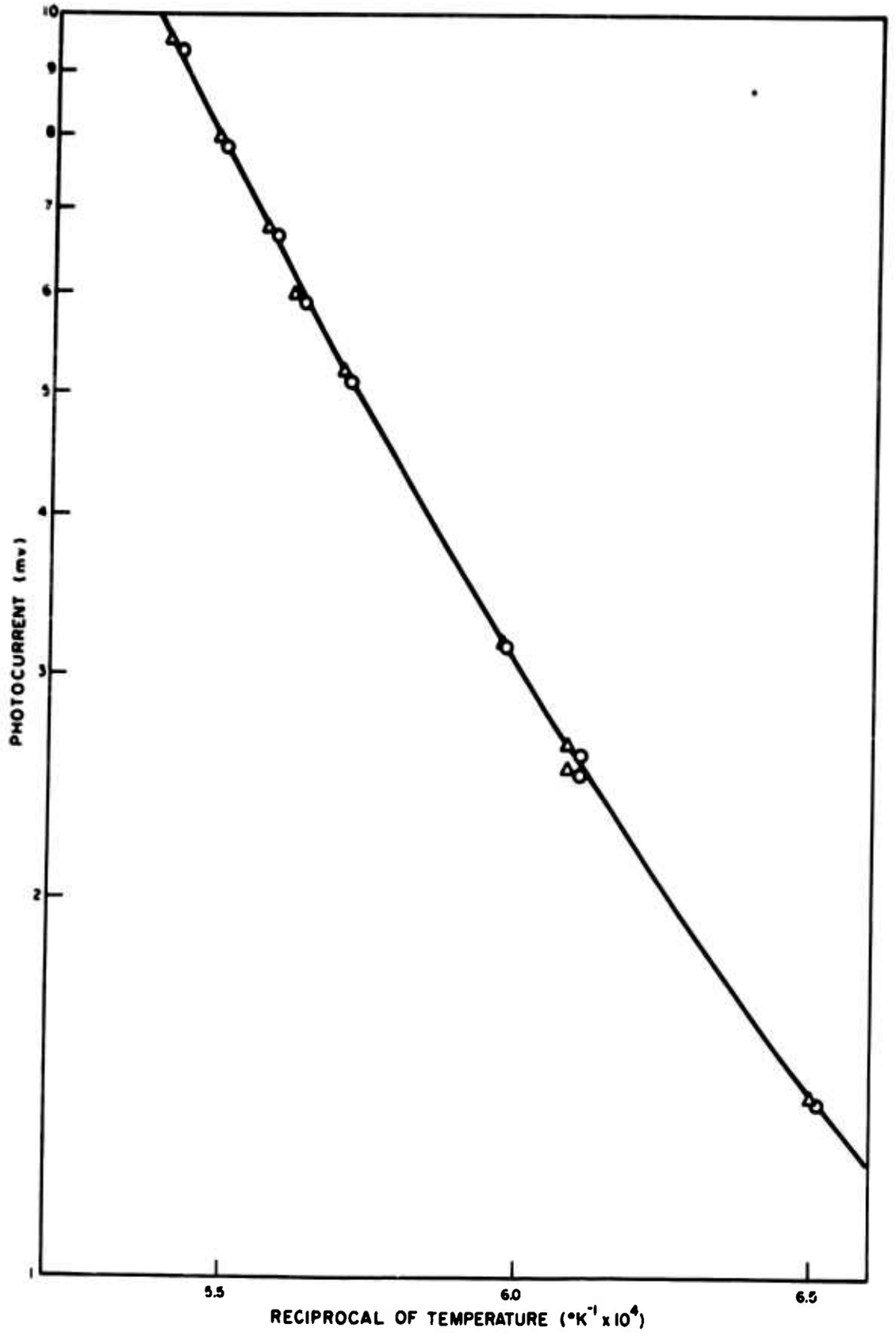

Figure 2. Calibration Curve of Photoelectric Pyrometer

Figure 3. Sample Holder for Specific Heat Measurements

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